

Review

A review of background dioxin concentrations in urban/suburban and rural soils across the United States: Implications for site assessments and the establishment of soil cleanup levels

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HIGHLIGHTS

- A review of available data to characterize background levels of dioxin-like compounds (DLCs) in U.S. soils.
- Background DLCs in urban/suburban soils were higher and more variable than in rural soils.
- Data indicate that background soil DLCs in urban areas may exceed regulatory remediation levels.

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abstract

Over the last several decades, dioxin releases have decreased by 90%, leading to a corresponding decrease in human body burdens. In addition, the weight-of-evidence indicates that soil exposures have little impact on human body burdens of dioxin-like compounds (DLCs), with dietary sources being the greatest contributor. In spite of this, USEPA recently proposed substantially lower preliminary remediation goals (PRGs) for soil based on their new oral reference dose (RfD) for dioxin. As such, it is important to understand how these lower soil PRGs compare to background concentrations in urban/suburban and rural soils. The objective of this evaluation was to conduct a comprehensive review of available data concerning background levels of DLCs in U.S. soils. There was substantial variability in how the soil dioxin data were presented (e.g., raw vs. summary data, congener vs. toxic equivalency [TEQ] concentration, number of DLC congeners reported, etc.). In cases where TEQ estimates were based on outdated TEFs and congener-specific data was provided, TEQ concentrations were recalculated using the current WHO₂₀₀₆ TEFs. The data available for rural soils were generally more robust than for urban/suburban soils. Not surprisingly, background levels of DLCs in urban/suburban soils were higher and more variable than in rural soils: 0.1–186 vs. 0.1–22.9 ng/kg TEQ, respectively. In several cases, incomplete soil DLC data were available (e.g., DL-PCBs not included) and, as such, calculated TEQ concentrations likely underestimate actual background levels. Though the current data are somewhat limited, these findings indicate that background DLC concentrations in urban/suburban soils may exceed the USEPA's updated PRGs based on the oral RfD, and are expected to substantially exceed future PRGs to be developed based on the forthcoming dioxin cancer slope factor. This demonstrates a need to characterize anthropogenic background DLCs in non-rural areas across the US to avoid establishing soil screening levels and PRGs that are lower than background concentrations.

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1. Introduction

Dioxin-like compounds (DLCs) are ubiquitous in soils due to the wide variety of sources that have contributed to background levels across the U.S. (USEPA, 2003). These sources have historically included a variety of natural and anthropogenic sources such as medical, municipal and hazardous waste combustion, combustion of fuels, backyard barrel burning, forest fires, volcanoes, as well as a variety of industrial sources such as secondary copper smelting, chlorine bleaching of paper and pulp, and the production of chlorinated phenols (USEPA, 2003). While much of the focus has historically been on industrial sources, in fact natural sources have been found to result in high levels of DLCs in soils as well. Deardorff et al. (2008) characterized PCDD/Fs in ash and topsoil following the 2007 wildfires in California, and reported that the highest levels were found in areas where homes were burned, followed by areas of agriculture and brush. Concentrations in the samples were found to range from 1.3 to 1680 ng/kg TEQ (based on WHO₂₀₀₆ TEFs). Importantly, environmental releases of DLCs decreased by approximately 90% between 1987 and 2000 (USEPA, 2006). These reductions were achieved by several means, including regulatory activities, improved emission controls, voluntary actions on behalf of industry, and the closing of a number of facilities. As a result of this decrease in dioxin emissions from industrial activities, activities such as forest and brush fires, backyard barrel burning, etc. are now the primary sources of emissions. Nonetheless, because DLCs are persistent in the environment, they are present in nearly all soils in the U.S. at varying concentrations depending on historical activities/sources in the region. As such, it is essential to understand and characterize background levels of DLCs in urban, suburban, agricultural, and rural soils across the U.S.

Dioxin-like compounds are associated with a number of different health effects, and therefore the characterization of background levels of DLCs in soils is particularly important for evaluating human health risk, especially given recent activity by the U.S. Environmental Protection Agency (USEPA). In December of 2009, the USEPA Office of Superfund Remediation and Technology Innovation released a guidance document titled Draft Recommended Interim Preliminary Remediation Goals for Dioxin in Soil at CERCLA and RCRA sites. In this guidance document the Agency proposed interim residential soil preliminary remediation goals (PRGs) for DLCs of 3.7 and 72 ng/kg TEQ, and interim commercial/industrial PRGs of 17 and 950 ng/kg TEQ, based on cancer and non-cancer endpoints, respectively. The Agency ultimately recommended setting the draft interim residential soil PRG at 72 ng/kg TEQ (down from a value of 1000 ng/kg TEQ that had been used for decades (USEPA, 1998)) and setting the draft interim commercial/industrial soil PRG at 950 ng/kg TEQ (down from a value of 5000–20,000 ng/kg TEQ that had been used for decades (USEPA, 1998)). The rationale

that the Agency provide for selecting these two values was that these PRGs "generally provide adequate protection against non-cancer effects," and in addition, "generally are protective for cancer effects at approximately the 1E-05 risk level, which is within USEPA's protective risk range of 1E-04 to 1E-06" (USEPA, 2009a). However, the USEPA never finalized their guidance document. Instead, following release of their final oral reference dose (RfD) for 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD), the Agency developed new residential and commercial/industrial PRGs based upon this final RfD and posted these PRGs on the Superfund Program's Non-Cancer Toxicity Value for Dioxin and CERCLA/RCRA Cleanups Question and Answer website (<http://www.epa.gov/superfund/health/contaminants/dioxin/dioxinsoil.html>). The new residential and commercial/industrial soil PRGs developed by USEPA based on this final RfD and default exposure parameters are 50 ng/kg TEQ and 664 ng/kg TEQ, respectively. These soil PRGs also appear on the USEPA Regional Screening Level (RSL) tables (although the commercial/industrial value has been rounded down to 600 ng/kg TEQ on the RSL tables). As already noted, the USEPA has yet to release their updated oral cancer slope factor (CSF) for TCDD, but it is anticipated – based on the draft oral CSF released for public comment in May of 2010 – that the final value will be substantially higher (more conservative) than the oral CSF used to develop both the proposed interim PRGs (i.e., 3.7 ng/kg) and the current cancer-based RSL (i.e., 4.5 ng/kg). Thus, any cancer-based PRG developed based on the new cancer slope factor (once released) is expected to be well below both of these previously calculated cancer-based PRGs.

Notably, in the proposed interim PRG document (USEPA, 2009a), the USEPA claimed that the proposed residential soil PRG was expected to be higher than typical background levels for residential soils. To support this claim, the Agency cited the 1998 ATSDR Toxicological Profile for Chlorinated Dibenzo-p-Dioxins (ATSDR, 1998). The ATSDR toxicological profile presents a general overview of the concentrations of DLCs in soils at National Priority List (NPL) sites, industrial, urban, and pristine rural sites in the U.S. The data are extremely limited and difficult to interpret as presented in the ATSDR document because much of the data are limited to TCDD only, are presented as homologues detected in soils without mention of concentration, are presented as absolute concentrations of homologue classes rather than as TEQ, do not account for dioxin-like polychlorinated biphenyls (DL-PCBs), or are presented as TEQ concentrations based on outdated toxicity equivalency factors (TEFs). These limitations likely result in underestimates of soil DLC levels, and make it extremely difficult to understand whether the interim PRGs are in fact above or below typical urban, suburban, agricultural, and rural background soil concentrations. As such, the objective of this assessment was to conduct a comprehensive review of all available published and unpublished data that could be obtained concerning the

levels of DLCs in soils in different types of areas, focusing in particular on TEQ concentrations where possible, and applying the most current TEFs developed by the World Health Organization (WHO) where feasible (van den Berg et al., 2006).

2. Methods

A comprehensive search was conducted in an attempt to identify both published and unpublished studies that reported concentrations of DLCs in background soils in the U.S. This involved searching the peer-reviewed published literature, state and federal regulatory websites, and relevant government documents, as well as a general search of the internet using common search engines. Any source that reported a concentration for TCDD or DLCs in background soils in the U.S. was obtained and reviewed. Primary sources were preferable, though if not available, data from secondary sources were also utilized. The first volume of the 2003 USEPA Dioxin Reassessment was a key secondary source as it summarized the background soils dioxin data available up to the time of its publication (USEPA, 2003). The comprehensive search identified a total of 18 studies with information on dioxin concentrations in background soils in the U.S. Importantly, only sample results from areas not believed to be impacted by recognized industrial sources of DLCs were retained for inclusion in this review. Primary sources were available for all but three of the studies (MRI, 1992; NIH, 1995; Tewhey Associates, 1997) and, as such, for these three, data presented in USEPA's 2003 Dioxin Reassessment were relied upon. Data were assumed to have been subject to appropriate data validation prior to publication in the peer-reviewed literature and/or in government reports. Independent data validation was not possible due to the lack of availability of analytical data packages in the publications/government reports.

The method for calculating and presenting dioxin soil concentrations often differed between the various studies, so an important component to this review was to translate these findings into common metrics (where possible), thereby allowing for direct comparisons. The most significant discrepancies among sources were the specific congeners analyzed in each of the studies, the availability of congener data for individual samples, method(s) used to impute values for non-detect results and/or the definitions of detection limits, and the TEF scheme (i.e., I-TEQ, WHO₁₉₉₈ TEF values, and WHO₂₀₀₆ TEF values) applied to calculate the total TEQ. While most studies reported results for polychlorinated dibenz-p-dioxin (PCDD) and polychlorinated dibenzofuran (PCDF) congeners, some studies also reported results for DL-PCBs, whereas other studies only reported results for TCDD. As a result, TEQ values across these studies do not represent a consistent set of DLCs, but rather a range of congeners.

In the analysis presented in this paper, data were categorized as representing either "urban/suburban" or "rural" soils. This categorization was determined on the following hierarchical criteria: 1) the original study authors explicitly designated a given data point as representing an urban or a rural location; 2) the original authors did not explicitly designate a given data point as either urban or rural but a designation to one of these two categories was made by the USEPA in the 2003 Dioxin Reassessment; or 3) if not explicitly designated as urban or rural by the original study authors or by the USEPA, then a judgment call was made based on the information provided in the study documentation. There were five studies that contained some data that did not have an explicit designation as either urban or rural (TOEQ, 1995a,b, 1997; RMCOEH, undated; Demond et al., 2008; HDOH, 2011). In these cases, information in the original study reports concerning sample locations was used to make a judgment call with regard to whether the sample reflected an urban/suburban or rural location. The specific judgments made are described in detail in the Results Section of study. Additionally, while none of the studies included an explicit designation of any data points as "suburban", because some of the samples were collected in areas designated as "residential" in the original study reports, and the descriptions suggested that these areas were more likely than not to be from

non-rural areas, these data were included in our "urban/suburban" category, along with data representing urban areas.

As a number of the study reports did not include individual sample results but instead only provided summary statistics (e.g., minimum, maximum, and/or mean), the data highlighted in this review are primarily limited to those same summary statistics. Median values are presented if these statistics were reported by the source, or if the data were available to calculate such. In instances where the total TEQ concentrations reported in the original study report reflected old TEF values (i.e., TEFs older than the current WHO₂₀₀₆ consensus-based TEF values) but individual congener data were provided, TEQ concentrations were recalculated using the WHO₂₀₀₆ TEF values. Brief study descriptions are provided in Table 1 for rural soils and Table 2 for urban/suburban soils. All soil TEQ concentrations are presented in units of nanograms/kilograms (ng/kg) on a dry weight basis.

3. Results

Our comprehensive research resulted in the identification of 18 studies with data on background concentrations of DLCs in soils in the United States, 14 of which contained data for rural soils and 11 of which contained data for urban/suburban soils. Data for rural soils reflected broad areas of the United States including Hawaii, the Northeast, Mid-Atlantic, South, Southwest, Pacific Northwest, and Midwest. Data for rural soils also includes a national soil survey that measured DLCs in soils from 27 rural sites across the continental U.S. (USEPA, 2007). In contrast, data for urban/suburban soils reflects fewer regions of the U.S.

In general, there are a number of methodological differences between the 18 studies including, but not limited to, the following: 1) differences in data reporting—some report data for individual congeners while others only report data for homolog groups or total TEQ; 2) differences in DLC congeners analyzed for—TCDD only, PCDD/Fs only, PCDD/Fs and DL-PCBs; 3) application of different TEF values to estimate TEQ concentrations (note—this is a function of the studies being conducted at different points in time); and 4) use of different proxy values for non-detected results in calculating the total TEQ concentration (e.g., the method detection limit (MDL) divided by √2, the full value of the MDL, zero, reporting limit (RL), MPC, etc.).

The minimum, maximum and mean TEQ concentrations determined for each of the 18 studies are presented in Tables 3 and 4 for rural and urban/suburban soils, respectively. Collectively, the data indicate that TEQ concentrations in background rural soils ranged from 0.1 to 22.9 ng/kg, while mean rural TEQ concentrations ranged from 1.1 to 7.1 ng/kg across the 14 studies that reported data for rural soils (Table 3). While rural mean concentrations were relatively low, 4 of the 14 studies had maximum concentrations over 20 ng/kg. As expected, the concentrations of DLCs in background urban/suburban soils were substantially higher and more variable than those in rural soils, with TEQ concentrations ranging from 0.1 to 186.2 ng/kg. The range of mean TEQ concentrations in urban/suburban soils was also substantially higher and ranged from 2.2 to 56.6 ng/kg. Importantly, 4 of the 11 studies with data for background urban/suburban soils reported maximum concentrations that exceed 100 ng/kg. Additional details regarding each of the 18 studies are presented below. (Note—studies are presented in chronological order.)

3.1. USEPA Midwest Soil Screening Survey (USEPA, 1985)

As a part of this survey, USEPA measured dioxin levels in soils in the following four Midwestern states: MI, IL, MN, and OH (USEPA, 1985). All samples were analyzed for TCDD, while a few samples were also analyzed for TCDF, OCDD, and OCDF. Samples from Henry, IL, Middletown, OH, and natural areas in Minnesota were collected from areas identified as rural by the USEPA (2003). Additionally, a select number of samples collected in MI were identified by the USEPA (2003) as being representative of "urban" soils. However, the survey report clearly indicates that

Table 1
Studies with information on background concentrations of DLCs in rural soils.

Source	Study/dataset description	Congener data reported	DLCs reported	Number of relevant samples	Detection limits reported
USEPA Midwest Soil Screening Survey (USEPA, 1985)	Soil screening study conducted in areas of the Midwest by the USEPA to define DLC levels in Midland, MI soils—USEPA Dioxin Reassessment identified soils collected in MN as representing rural.	YES	Mostly TCDD, some TCDF, OCDD, OCDF	4	YES
Midwest and Ontario Soil Survey (Birmingham, 1990)	As a part of a human exposure study, DLC levels were measured in U.S. Midwestern and Canadian soils from various land use areas, including rural.	NO	PCDD/F	30	NO
Elk River, Minnesota Survey (Reed et al., 1990)	A baseline survey of DLC levels in soils from the vicinity of an area along the Elk River in MN at the site of an refuse-derived fuel-powered electric generation station—background soils were identified as those collected prior to the facility operation.	YES	PCDD/F	4	YES
Connecticut State Survey (MRI, 1992)	USEPA Dioxin Reassessment identified a Connecticut soil survey wherein soil samples were collected from cities throughout Connecticut to evaluate the impact of municipal waste incinerators on DLC levels in soils—soils from pre-operational sites were identified as background rural soils.	YES	PCDD/F	34	NO
Mississippi Rural survey (Fiedler et al., 1995; Rappe et al., 1997)	Monitoring program performed to determine baseline concentrations of DLCs in rural areas across 8 counties in southern Mississippi.	NO	PCDD/F	36	For TCDD only
TCEQ Texas Soil Survey (TCEQ, 1995a,b)	Survey was conducted by the state of Texas to characterize DLC levels in soils near municipal solid waste or medical waste incinerators in 5 cities (left, Carthage, Cleburne, Terrell, Quitman, and Midlothian)—areas outside of the city limits were considered to represent background rural soils.	YES	PCDD/F	16	NO
USEPA Columbus, Ohio (USEPA, 1996)	Background soils from within the city of Columbus and a rural area 3 miles outside of the city were collected to assess the impact of the city's Waste-to-Energy municipal incinerator on DLC levels in soils near the plant—soil samples collected 3 miles from the city were identified as rural.	YES	PCDD/F	3	YES
Maine soil survey (Tewhey Associates, 1997)	USEPA Dioxin Reassessment identified a rural soil survey wherein background soil samples were collected from the Yarmouth Pole Yard Site located in Yarmouth, Maine.	YES	PCDD/F	8	NO
TCEQ Texas Soil Survey (TCEQ, 1997)	Survey was conducted by the state of Texas to characterize DLC levels in soils near municipal solid waste or medical waste incinerators in 5 cities (Amarillo, Terrell, left, Pearland, and Galveston)—areas outside of the city limits were considered to represent background rural soils.	YES	PCDD/F	8	NO
Denver, Colorado Urban and Rural Background Soil Survey (USEPA, 2002)	DLC concentrations measured in diverse soil types in Denver metropolitan area to serve as background comparison for soils collected at Rocky Mountain Arsenal—soil samples from agricultural and open space areas represented rural soils.	YES	PCDD/F & PCBs	64	YES
USEPA National Rural Background Dioxin Soil Sample Survey (USEPA, 2007)	Pilot survey conducted by USEPA to characterize levels of DLCs in rural soil samples collected from several locations across the United States.	YES	PCDD/F & PCBs	27	NO
Washington State Department of Ecology study (WDE, 2010)	Survey was conducted to determine the background levels of metals and dioxins in Washington state soils—USEPA Dioxin Reassessment classified open space and forested soils as representing rural soils.	YES	PCDD/F	16	YES
Washington State Department of Ecology study (WDE, 2011a)	Follow up rural survey performed to re-evaluate rural background contaminants in forest and open range soils collected in Washington State parks.	YES	PCDD/F	41	YES
Hawaii Soil Dioxin Contamination Report (HDOH, 2011)	Dioxin-like compound levels in soils from non-sugarcane operations areas provided to compare with DLC level data for pesticide-related contamination of soils associated with former sugarcane operations throughout the State of Hawaii.	NO	PCDD/F	na	NO

NA = data on the number of samples analyzed at each of the 3 sites were not available.

□ Rural soils were not explicitly designated in study or by USEPA (2003); authors made a judgment call.

the Henry, IL and Middletown, OH samples were collected from areas considered to be more consistent with an urban setting. There were a total of 35 soil samples collected in Henry, IL and Middletown, OH, where TCDD levels were reported to range from non-detect to 5 ng/kg. For the limited number of the Middletown, OH samples, TCDF, OCDD, and OCDF congeners were also analyzed, yielding absolute (non-TEF-adjusted) concentrations ranging from non-detect to 6 ng/kg for TCDF, from 170 to 10,600 ng/kg for OCDD, and from non-detect to 50 ng/kg for OCDF. No specific source of OCDD was identified for the Ohio samples. Further, the samples in Middletown were collected in areas upwind of any potential industrial sources (USEPA, 1985). TCDD, TCDF, and OCDF were not detected in any of the samples collected from the areas in Minnesota designated as natural (i.e., areas away from roadways), although OCDD ranged from 92 to 200 ng/kg. Since data were available for each of these four congeners on an individual basis, the WHO₂₀₀₆ TEFs were applied to the absolute concentrations

presented in USEPA (1985) to estimate the TEQ₂₀₀₆ concentration for these four congeners for relevant samples. Based on this update, the TEQ₂₀₀₆ concentration was found to range from 2.2 to 7.6 ng/kg TEQ₂₀₀₆ for the Ohio soils (mean = 4.1 ng/kg TEQ₂₀₀₆), and 1.3–3.2 ng/kg (mean = 2.2 ng/kg TEQ₂₀₀₆) for the Minnesota soils for these 4 congeners.

Although the USEPA classified a subgroup of the 23 MI soil samples as representative of "urban" background, these urban soil samples were collected within the city of Midland, an area with a known dioxin source and, as such, they may not be representative of "typical" urban background (Nestrick et al., 1986). In the subgroup of urban soil samples, TCDD concentrations ranged from 3 to 170 ng/kg (mean of 56.5 ng/kg). For the limited number of Midland samples that were also analyzed for TCDF, OCDD, and OCDF, absolute (non-TEF-adjusted) concentrations were found to range from non-detect to 15 ng/kg for TCDF, from 680 to 12,000 ng/kg for OCDD, and from 44 to 669 ng/kg

Table 2

Studies with information on background concentrations of DLCs in urban/suburban soils.

Source	Study/dataset description	Congener data reported	DLCs reported	Number of relevant samples	Detection limits reported
USEPA Midwest Soil Screening Survey (USEPA, 1985)	Soil screening study conducted in areas of the Midwest by the USEPA to define DLC levels in Midland, MI soils—USEPA Dioxin Reassessment identified soils collected in public use areas of Midland, MI representing urban.	YES	Mostly TCDD, some TCDF, OCDD, OCDF	9	YES
USEPA Midwest Soil Screening Survey (USEPA, 1985)	Soil screening study conducted in areas of the Midwest by the USEPA to define DLC levels in Midland, MI soils—although USEPA Dioxin Reassessment identified background soils collected in areas of Henry, IL and Middleton, OH as rural, they were reclassified as urban for the current study since they were collected in residential and public areas of these small cities.	YES	Mostly TCDD, some TCDF, OCDD, OCDF	35	YES
Survey of Midwestern and Mid-Atlantic Cities (Nestrick et al., 1986)	A survey of DLC levels in soils in the vicinity of urban industrial areas in several Midwestern and Mid-Atlantic cities was conducted to provide background soil data for Midland, MI soils.	YES	TCDD only	20	YES
Midwest and Ontario Soil Survey (Birmingham, 1990)	As a part of a human exposure study, DLC levels were measured in U.S. Midwestern and Canadian soils from various land use areas, including urban.	NO	PCDD/F	47	NO
NIH campus soil survey in Bethesda, MD (NIH, 1995)	USEPA Dioxin Reassessment identified an urban soil survey by the National Institutes of Health (NIH) performed to determine the effect of 30 years of pathological waste incineration on the NIH campus and its surroundings.	YES	PCDD/F	37	NO
USEPA Columbus, Ohio (USEPA, 1996)	Background soils from within the city of Columbus and a rural area 3 miles outside of the city were collected to assess the impact of the city's Waste-to-Energy municipal incinerator on DLC levels in soils near the plant—soil samples from parts of the city away from the incinerator were identified as urban.	YES	PCDD/F	17	YES
TCEQ Texas Soil Survey (TCEQ, 1997)	Survey was conducted by the state of Texas to characterize DLC levels in soils near municipal solid waste or medical waste incinerators in 5 cities (Amarillo, Terrell, left, Pearland, and Galveston)—samples collected in the city of Austin represented background urban soils.	YES	PCDD/F	2	NO
Denver, CO soil survey (USEPA, 2002)	DLC concentrations measured in diverse soil types in Denver metropolitan area to serve as background comparison for soils collected at Rocky Mountain Arsenal site—USEPA Dioxin Reassessment classified soil samples from residential, commercial, and industrial areas as representing urban soils.	YES	PCDD/F & PCBs	98	YES
University of Utah Davis County Residential Soil Study (RMCOEH, undated)	Investigators with the University of Utah's Rocky Mountain left for Occupational and Environmental Health collected surface soil samples from residential properties throughout Davis County, Utah, and compared PCDD/F analytical results with the residential surface soils reported in USEPA's Denver Front Range study.	NO	PCDD/F	22	YES
University of Michigan Dioxin Exposure Study (Demond et al., 2008) (surface soil) ^a	Soil samples collected from residential plots in Jackson and Calhoun Counties of Michigan to serve as background soil dioxins for city of Midland, Michigan.	YES	PCDD/F & PCBs	194	NO
Washington State Department of Ecology study (WDE, 2010)	Survey was conducted to determine the background levels of metals and dioxins in Washington state soils—soils from several urban lefts in Washington were collected.	YES	PCDD/F	14	YES
Washington State Department of Ecology study (WDE, 2011b)	Follow up urban survey performed to re-evaluate urban background dioxin and PAH contaminants in soils collected in six areas around Seattle, WA.	YES	PCDD/F	120	YES

^a Urban soils were not explicitly designated in study or by USEPA (2003); authors made a judgment call.

for OCDF. The elevated OCDD levels detected in the Midland samples were attributed to emissions from industrial chemical waste incineration (USEPA, 1985). Applying the WHO₂₀₀₆ TEFs to absolute concentration data for the individual congeners yielded a mean TEQ₂₀₀₆ of 51.2 ng/kg based on individual sample results ranging from 4.1 to 112 ng/kg TEQ₂₀₀₆ for the samples where TCDD, TCDF, OCDD, and OCDF were determined.

3.2. Midwestern and Mid-Atlantic Cities Soil Survey (Nestrick et al., 1986)

Nestrick et al. (1986) measured the levels of dioxins in background soils in 16 cities in the Midwest and Mid-Atlantic regions of the U.S. As a part of this study, 20 soils samples were collected within one-mile of major steel, automotive, or chemical manufacturing facilities, or municipal solid waste incinerators. The study authors identified these soils as representing urban background since the potential sources were believed to be common to urban settings. The investigatorsonly analyzed

samples for TCDD, and concentrations were reported to range from the lowest detection limit (0.2 ng/kg TCDD) to 9.4 ng/kg, with a mean of 2.2 ng/kg. This study was the only one evaluated that only looked at the TCDD, and, as such, these concentrations are considered to be biased low and of limited use in defining typical urban background concentrations, given the lack of data on all of the other DLCs.

3.3. Elk River, Minnesota Soil Survey (Reed et al., 1990)

The Minnesota Pollution Control Agency (MPCA) collected soil samples in an area near the Elk River in MN where an electric generating station was to be constructed, and analyzed those samples for PCDDs/Fs. The objective of this study was to collect baseline data on environmental media prior to the electric generating station coming online. The Elk River area was an agricultural area with no major industrial or commercial activities at the time of the survey, the soils samples of which were interpreted by the USEPA (2003) as representative of rural soils. The

Table 3
Background concentrations of DLCs in rural soils.

Source	Source data available for TEQ calculation/recalculation?	TEF	Imputed value for non-detect	Mean TEQ (ng/kg)	Median TEQ (ng/kg)	Minimum TEQ (ng/kg)	Maximum TEQ (ng/kg)
TCEQ Texas Soil Survey (TCEQ, 1997) [†]	YES	2006 WHO	Full MPC	7.1	4.1	0.8	22.6
Connecticut State Survey (MRI, 1992)	NO	1998 WHO	Zero	5.7	NR	NR	NR
Elk River, Minnesota Survey (Reed et al., 1990)	NO	2006 WHO	Zero	3.9	3.8	0.6	7.6
Mississippi Rural Survey (Fiedler et al., 1995; Rappe et al., 1997) [†]	YES	2006 WHO	NR	3.1	0.8	0.1	22.9
TCEQ Texas Soil Survey (TCEQ, 1995a,b) [†]	YES	2006 WHO	Full MPC	2.9	2.3	0.8	8.0
Maine soil survey (Tewhey Associates, 1997)	NO	1998 WHO	Zero	2.9	NR	NR	NR
Washington State Department of Ecology study (WDE, 2010) [†]	YES	2006 WHO	DL/2	2.5	1.9	0.7	6.6
Denver, Colorado Urban and Rural Background Soil Survey (USEPA, 2002) [†]	YES	2006 WHO	DL/2	2.2	1.1	0.1	20.2
USEPA Midwest Soil Screening Survey (USEPA, 1985) [†]	YES	2006 WHO	Full DL	TCDD-only: 2 All congeners: 2.2	TCDD-only: 2 All congeners: 2.1	TCDD-only: 1 All congeners: 1.3	TCDD-only: 3 All congeners: 3.2
USEPA National Rural Background Dioxin Soil Sample Survey (USEPA, 2007) [†]	YES	2006 WHO	RL/2	2.0	1.0	0.2	11.8
Washington State Department of Ecology study (WDE, 2011a)	YES	2006 WHO	DL/2	1.7	0.6	0.1	9.4
USEPA Columbus, Ohio (USEPA, 1996)	YES	2006 WHO	DL/2	1.3	1.2	1.0	1.8
Midwest and Ontario Soil Survey (Birmingham, 1990)	NO	1989 USEPA	DL/2	0.4	0.2	0.2	2.2
Hawaii Soil Dioxin Contamination Report (HDOH, 2011)	NO	2006 WHO	NR	NR	NR	0.9	27.0

MPC = maximum possible concentration.

DL = detection limit.

RL = reporting limit.

NR = not reported.

[†] Updated TEQ concentrations were derived for this paper using WHO₂₀₀₆ TEFs.

authors provided congener data for the four soil samples collected, but did not derive TEQ-based concentrations. The USEPA, however, calculated a mean TEQ concentration of 3.6 ng/kg using the WHO₁₉₉₈ TEFs (USEPA, 2003). Since the congener data were available, the total

TEQ concentrations for these four samples were updated using the WHO₂₀₀₆ TEFs. Based on this update, TEQ₂₀₀₆ concentrations were found to range from 0.6 ng/kg to 7.6 ng/kg, with a mean TEQ concentration of 3.9 ng/kg TEQ₂₀₀₆.

Table 4
Background concentrations of DLCs in urban/suburban soils.

Source	Source data available for TEQ calculation/recalculation?	TEF	Imputed value for non-detect	Mean TEQ (ng/kg)	Median TEQ (ng/kg)	Minimum TEQ (ng/kg)	Maximum TEQ (ng/kg)
USEPA Midwest Soil Screening Survey (USEPA, 1985) [†]	YES	2006 WHO	NR	TCDD-only: 56.6 All congeners: 51.2	TCDD-only: 28.0 All congeners: 48.4	TCDD-only: 3.0 All congeners: 4.1	TCDD-only: 170.0 All congeners: 112.0
USEPA Columbus, Ohio (USEPA, 1996) [†]	YES	2006 WHO	NR	20.2	9.2	3.0	64.0
Washington State Department of Ecology study (WDE, 2011b)	YES	2006 WHO	DL/2	19.2	12.0	1.7	120
Denver, CO soil survey (USEPA, 2002) [†]	YES	2006 WHO	DL/2	13.1	4.4	0.2	145.7
Midwest and Ontario Soil Survey (Birmingham, 1990)	NO	1989 USEPA	DL/2	9.4	1.2	0.1	78.5
University of Michigan Dioxin Exposure Study (Demond et al., 2008) (surface soil)	NO	2006 WHO	DL/2	6.9	3.6	0.4	186.2
TCEQ Texas Soil Survey (TCEQ, 1997) [†]	YES	2006 WHO	Full MPC	6.7	6.7 [†]	6.2	7.2
Washington State Department of Ecology study (WDE, 2010) [†]	YES	2006 WHO	DL/2	4.2	2.0	0.7	21.0
USEPA Midwest Soil Screening Survey – Henry, IL and Middleton, OH (USEPA, 1985) [†]	YES	2006 WHO	NR	TCDD-only: 2.0 All congeners: 4.1	TCDD-only: 2.0 All congeners: 3.3	TCDD-only: 1.0 All congeners: 2.2	TCDD-only: 5.0 All congeners: 7.6
NIH campus soil survey in Bethesda, MD (NIH, 1995)	NO	1998 WHO	Zero	2.2	NR	NR	NR
Survey of Midwestern and Mid-Atlantic Cities (Nestrick et al., 1986)	YES	TCDD only	Full DL	2.2	0.9	0.2	9.4
University of Utah (RMCOEH, undated)	NO	1998 WHO	DL/2	1.3	0.9	0.3	4.5

MPC = maximum possible concentration.

DL = detection limit.

NR = not reported.

[†] Updated TEQ concentrations were derived for this paper using WHO₂₀₀₆ TEFs.

[†] This dataset is comprised of 2 samples, so median = mean.

3.4. Midwest and Ontario Soil Survey (Birmingham, 1990)

Birmingham (1990) published results on soil samples collected in the Midwestern U.S. and Ontario, Canada and reported TEQ concentrations for several soil samples designated as either rural ($n = 30$) or urban ($n = 47$) by the original study authors. The authors did not distinguish between the Canadian and U.S. samples and, as such, the data could not be broken out to reflect U.S.-only background concentrations. The study authors analyzed for individual PCDD/F congeners and then combined the congeners into homologue groups and applied the I-TEFs to homologue data (rather than to individual congener data, as is typically done) to estimate a TEQ concentration for each sample. While this is not ideal, the overall impact is likely negligible because the I-TEFs for individual congeners within each homologue class are the same except for the two penta-furans, which were not detected in any of the rural samples and were detected very infrequently in the urban samples. Based on this analysis, I-TEQ concentrations were reported to range from 0.2 to 2.2 ng/kg in rural soils (mean 1.1 ng/kg I-TEQ) and 0.1 to 78.5 ng/kg in urban soils (mean 9.4 ng/kg I-TEQ). The application of TEFs to DLC homolog group concentrations does not comport with the currently accepted method for deriving TEQ concentrations (van den Berg et al., 1998, 2006). However, individual congener data were not available and, as such, it was not possible to re-calculate the concentrations using the WHO₂₀₀₆ TEFs.

3.5. 1995 TCEQ Texas Soil Survey (TCEQ, 1995a,b)

In response to concerns about the potential for municipal solid waste and medical waste incinerators to release PCDDs/Fs, the TCEQ conducted soil sampling in six east and central Texas rural communities where such facilities were operating (TCEQ, 1995a,b). In addition to collecting samples in the vicinity of the incinerators, two to six background samples were collected in areas away from any recognized dioxin point source in each community. A total of 16 background soil samples were collected. Although the Agency did not explicitly identify samples as either urban or rural, the sample location coordinates provided in the report were used to classify the samples. Based on this evaluation, it was determined that all of the background samples in this study were representative of rural soils. In a 1995 TCEQ memo summarizing the results of this study, the Agency reported that concentrations in these background soils ranged from 0.8 to 7.3 ng/kg I-TEQ. As the individual congener data were available, the total TEQ concentrations for each sample were updated based on the WHO₂₀₀₆ TEFs. Based on this update, total TEQ₂₀₀₆ was found to range from 0.8 to 8.0 ng/kg TEQ₂₀₀₆, with a mean of 2.9 ng/kg TEQ₂₀₀₆.

3.6. Mississippi Rural Soil Survey (Fiedler et al., 1995)

Fiedler et al. (1995) measured dioxin levels in soils in rural areas across 8 counties in Mississippi. The authors calculated a mean I-TEQ of 3.1 ng/kg for PCDD/Fs, based on concentrations ranging from 0.1 to 22.9 ng/kg I-TEQ across all 36 samples. Rappe et al. (1997) also published results based on this data set and provided sample specific information for TCDD and OCDD, although there was no information on how TEQ concentrations were derived. In addition, there are slight variations in sample TEQs between the two publications. Because Rappe et al. (1997) did not identify which TEQ methodology they employed, only the results from Fiedler et al. (1995) are presented herein.

3.7. USEPA Columbus, Ohio Waste-to-Energy Municipal Incinerator Dioxin Soil Sampling Project (USEPA, 1996)

The USEPA collected samples in both urban and rural areas in and around Columbus, OH (USEPA, 1996) as part of an investigation to evaluate PCDD/PCDF soil concentrations in the vicinity of the Columbus

Waste-to-Energy facility. The samples were explicitly designated as either urban or rural by the USEPA (USEPA, 1996, 2003). For the three rural samples, the USEPA reported a mean TEQ concentration of 1.4 ng/kg based on application of the I-TEFs (min-max: 1–2 ng/kg I-TEQ). For the eighteen urban samples, the USEPA reported a mean of 19 ng/kg I-TEQ (min–max: 2.8–61.3 ng/kg I-TEQ). As individual congener data were provided in the report for all three rural, and 17 of the 18 urban, soil samples, the TEQ concentrations were updated based on the WHO₂₀₀₆ TEFs. Based on this update, TEQ₂₀₀₆ concentrations were found to range from 1.0 to 1.8 ng/kg (mean 1.3 ng/kg) in rural soils and from 3.0 to 64.0 ng/kg (mean 20.2 ng/kg) in urban samples.

3.8. 1997 TCEQ Texas Soil Survey (TCEQ, 1997)

The TCEQ conducted an initial dioxin soil survey in 1995 (described above). In 1997 the Agency conducted a follow up study to look at an additional five Texas communities in an attempt to characterize a larger portion of the state (including northern and southeastern Texas) (TCEQ, 1997). Though not explicitly characterized as such by the original study authors, these samples were judged to represent rural soils based on the sample location coordinates provided in the study report. In addition, this study also included two background samples that the original study authors stated were collected "at sites which were chosen to represent typical urban background." A total of eight rural and two urban samples were collected as a part of this follow up soil survey. In a memorandum summarizing the results of this follow up study (TCEQ, 1997), the Agency reported that rural background soil dioxin levels ranged from 0.6 to 23.1 ng/kg I-TEQ, while the urban soil samples collected in Austin, TX ranged from 4.1 to 5.3 ng/kg I-TEQ. As individual congener data were provided in the report, the TEQ concentrations were updated based on the WHO₂₀₀₆ TEFs. Based on this update, TEQ₂₀₀₆ concentrations were found to range from 0.8 to 22.6 ng/kg (mean 7.1 ng/kg) in rural soils and from 6.2–7.2 ng/kg (mean 6.7 ng/kg) in urban samples.

3.9. USEPA Denver, Colorado Urban and Rural Background Soil Survey (USEPA, 2002)

USEPA Region 8 conducted a comprehensive survey of dioxin concentrations in background soils as a part of an investigation at the Rocky Mountain Arsenal site near Denver, CO (USEPA, 2002). More than 170 soil samples were collected from "off-site" areas on public lands and analyzed for PCDD/Fs and DL-PCBs, though only 160 samples were used to derive summary statistics. These samples were collected from agricultural, open space, residential, commercial, and industrial areas, and were considered by the USEPA to represent background since they were not associated with point sources. The agricultural and open space areas were classified as rural by the original study authors. The remaining sample types (i.e., residential, commercial, and industrial), while not explicitly designated as urban/suburban by the original study authors, were classified as urban by the USEPA (USEPA, 2003) and, as such, were included in the urban/suburban category for purposes of this current assessment.

The total TEQ concentrations provided in the original study report were based on the WHO₁₉₉₈ TEFs. However, since individual congener data were provided in the study report, the total TEQ concentrations were updated to reflect the WHO₂₀₀₆ TEFs. It should be noted that, in their original analysis, the USEPA excluded two samples (i.e., one residential sample with a total TEQ₂₀₀₆ concentration of 145 ng/kg and one commercial sample with a total TEQ₂₀₀₆ of 146 ng/kg) based on the fact that they fell outside of the 99th percentile of the dataset. The data for these two soil samples were included in the updated calculations for the urban/suburban area soils in this current assessment because it was determined that they had been inappropriately excluded.

from the earlier analysis based on an unorthodox outlier identification approach (simple dataset percentile comparison with a non-standard cut-off). Additionally, as noted by USEPA, no evidence was observed, either at the sampling sites or based on the congener profiles themselves, to suggest that either sample was influenced by some point source of contamination (USEPA, 2002). Based on this update, TEQ₂₀₀₆ concentrations were found to range from 0.1 to 20.2 ng/kg in rural soils and from 0.2 to 146 ng/kg in urban/suburban soils.

This study is the most comprehensive study of background concentrations performed to date based on the number of samples and the types of land-uses sampled. For this reason, additional analyses were conducted to assess individual congener and class (i.e., PCDDs, PCDFs, and DL-PCBs) contributions to total TEQ₂₀₀₆ for each of the five distinct land-use areas. Based on this analysis, it was determined that on average, PCDDs contributed to at least half of total TEQ₂₀₀₆ in all cases, regardless of land use type (Fig. 1). PCDD contributions to total TEQ₂₀₀₆ were greatest for residential and commercial soils (60% and 62%, respectively), while PCDF contributions were greatest for agricultural soils (29%) and DL-PCB contributions were greatest for industrial and open space soils (27% and 25%, respectively). PCDFs and DL-PCBs were equal contributors to the total TEQ₂₀₀₆ in the residential and commercial soils, while DL-PCBs comprised a slightly greater portion (7%) of total TEQ₂₀₀₆ than did PCDFs in industrial soils. The DLC congener TEQ profiles were dominated by PCB126, 1,2,3,7,8-PeCDD, and 1,2,3,4,6,7,8-HxCDD, regardless of soil type (Fig. 2). Together, the average contributions of these three congeners comprised more than 50% of the total TEQ₂₀₀₆ for all land uses. For the rural soils (i.e., agricultural and open space soils), 2,3,4,7,8-PeCDF was the next highest contributor to total TEQ, followed by 1,2,3,6,7,8-HxCDD. TCDD – the most potent of the DLC congeners – was a more significant contributor to total TEQ levels in the residential and commercial soils, accounting for 7% and 9% of the total TEQ₂₀₀₆, respectively (Fig. 2). Interestingly, TCDD was a less significant total TEQ₂₀₀₆ contributor in industrial soils than the other urban/suburban soil types or even agricultural soils. From a broader perspective, similar findings were observed when examining the congener contributions to total background TEQ in residential soils from Jackson/Calhoun counties (Michigan) — that is, the authors observed the same congeners contributing most to total TEQ (Demond et al., 2008). These similarities suggest that similar natural and/or anthropogenic sources are likely contributing to ubiquitous background concentrations in soils in across the U.S.

3.10. University of Utah Davis County Residential Soil Study (RMCOEH, undated)

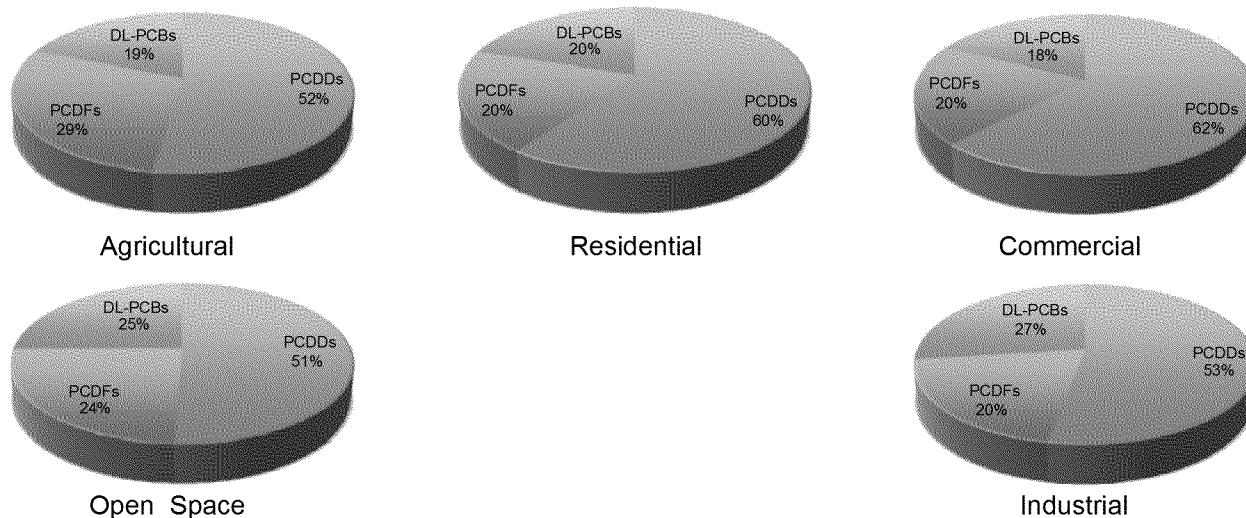
Investigators with the University of Utah's Rocky Mountain Center for Occupational and Environmental Health evaluated surface soil samples from public properties adjacent to residential developments throughout Davis County, Utah. The study was conducted to characterize baseline soil dioxin levels to facilitate evaluation of potential contamination in other areas. Though undated, the study appears to have been conducted between 2002 and 2006 since the report compares the Davis County residential soil results with those reported by USEPA in the 2002 Denver, Colorado Front Range report, but does not utilize nor mention the most recent set of consensus WHO TEFs published by van den Berg et al. (2006). The authors of the University of Utah report followed the approach used by the USEPA for the Denver, Colorado study, with comparable data flags and the utilization of one-half detection limit values for undetected PCDD/F analytes. However, the Davis County soil study was limited to the 17 PCDD/F congeners, and did not include the 12 DL-PCB congeners. The paper summarizing the results of this study reported that concentrations in these background soils ranged from 0.32 to 4.47 ng/kg TEQ₁₉₉₈. As individual congener data were not available, it was not possible to re-calculate the concentrations using the WHO₂₀₀₆ TEFs.

3.11. USEPA National Rural Background Dioxin Soil Sample Survey (USEPA, 2007)

The USEPA conducted a survey of PCDD/Fs and DL-PCBs in rural soils across the United States (USEPA, 2007). One soil sample was collected from each of 27 different rural sites across the country and TEQ concentrations were derived based on the WHO₁₉₉₈ TEFs. All sample locations were explicitly designated as rural by the original study authors. In addition, the report provided the analytical data for each DLC congener, and, as such, the TEQ concentrations were updated to reflect the WHO₂₀₀₆ TEFs. Based on these updates, total TEQs were found to range from 0.3 to 11.8 ng TEQ₂₀₀₆/kg dry soil, with a mean of 2.0 ng TEQ₂₀₀₆/kg dry soil.

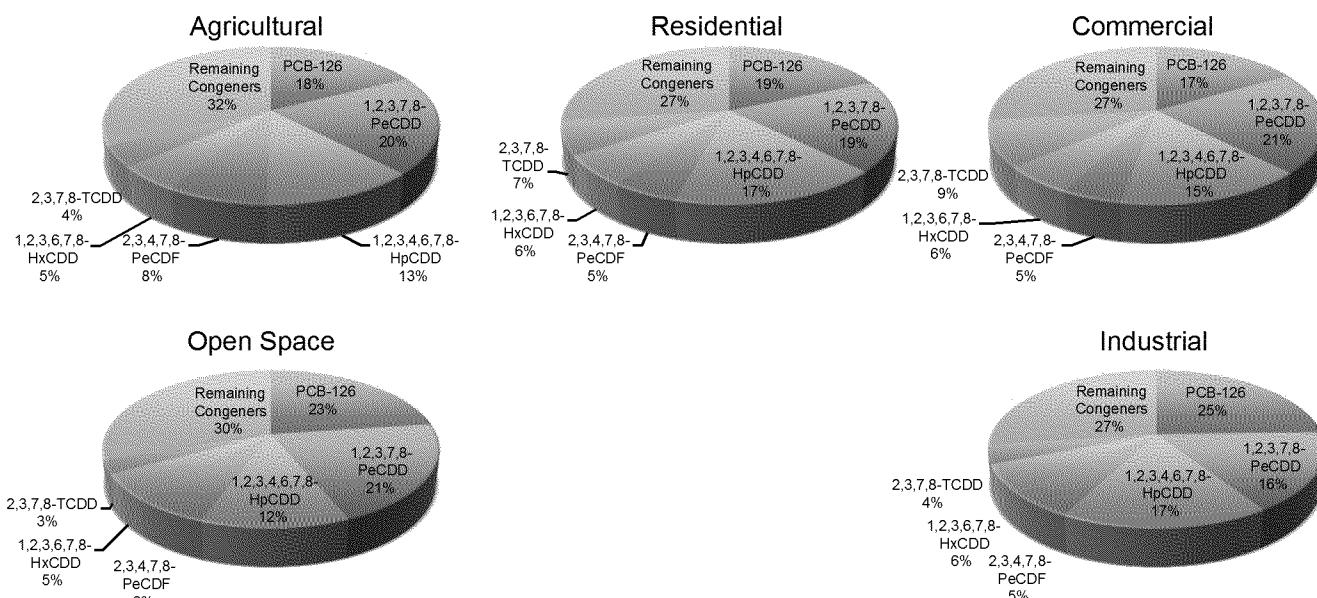
3.12. University of Michigan Dioxin Exposure Study (UMDES) (Demond et al., 2008)

The UMDES was conducted in an effort to characterize dioxin concentrations in several media (soils, house dust, human serum) in and around



†Data from 2002 USEPA Denver Urban and Rural Background Soil Survey.

Fig. 1. Contribution of PCDDs, PCDFs, and DL-PCBs to Background Soil Total TEQ. Data from 2002 USEPA Denver Urban and Rural Background Soil Survey.



†Data from 2002 USEPA Denver Urban and Rural Background Soil Survey.

Fig. 2. Contribution of Individual Congeners to Background Soil Total TEQ ($\geq 5\%$ Contribution + TCDD). Data from 2002 USEPA Denver Urban and Rural Background Soil Survey.

the city of Midland, Michigan, as well as in a control community in Jackson and Calhoun Counties in Michigan (Demond et al., 2008). The samples collected in the control community were identified as representing a mix of rural and urban/suburbansites. However, neither individual sample results nor sample locations were provided in any of the UMDES reports or publications. Based on descriptions provided about the control community and the focus on collecting samples in residential areas, the samples collected in the control community were assumed to be more representative of urban/suburban for the purposes of this evaluation. The original study authors reported that concentrations of PCDD/Fs and DL-PCBs in 0–1 inch house perimeter samples in the control communities ranged from 0.4 to 186.2 ng TEQ₂₀₀₆/kg soil (mean of 6.9 ng/kg), while concentrations in 1–6 inch house perimeter samples ranged from 0.9 to 35.3 ng/kg (mean of 11.2 ng/kg). However, it is important to note that if the TEQ concentration was based solely on PCDD/F congeners (and excluded DL-PCBs), then the maximum background TEQ concentration in the 0–1 inch house perimeter samples in the UMDES control community was 64.1 ng/kg. This suggests that there may have been an unidentified PCB source present at the location of the 0–1 inch house perimeter sample having a total TEQ of 186.2 ng/kg, and, therefore, this sample may not be representative of an actual background TEQ concentration.

3.13. Washington State Department of Ecology (WDE) Study (WDE, 2010)

WDE measured dioxin concentrations in soils in open range, forest, and urban areas in the state of Washington to provide an initial assessment of typical concentrations of dioxins in Washington soils. Fourteen urban, eight open range, and eight forest soil samples were collected and analyzed, of which the latter two soil types were classified by USEPA (2003) as representing rural soils. The investigators applied the I-TEFs to estimate a total TEQ for each sample, and reported mean TEQ concentrations of 4.1 (min–max range: 0.1–19), 1.0 (min–max range: 0.04–4.6), and 2.3 (min–max range: 0.03–5.2) ng/kg for the urban, open range and forest samples, respectively. This work was originally published by Rogowski et al. (1999), and no mention was made of the history or potential impact of forest fires on the TEQ concentrations measured in the background forest soil samples. More recently, the WDE went back and applied the WHO₂₀₀₆ TEFs to the soils data, and

calculated a mean rural background soil TEQ₂₀₀₆ concentration of 2.5 ng/kg using the open range and forest soil sample data, based on a range of 0.7–6.6 ng/kg TEQ₂₀₀₆. The urban dataset was also updated, and the study authors reported a mean urban background soil TEQ₂₀₀₆ concentration of 4.2 ng/kg TEQ₂₀₀₆ based on a range of 0.7–21 ng/kg TEQ₂₀₀₆ (WDE, 2010).

3.14. Washington State Department of Ecology (WDE) Study (WDE, 2011a,b)

In a follow up to their earlier effort (WDE, 2010), the WDE conducted two separate and more comprehensive soil dioxin surveys in samples from rural (WDE, 2011a) and urban/suburban (WDE, 2011b) areas in the state of Washington. The rural samples represent a combination of soils from forested and open range areas of several Washington state parks ($n = 41$), whereas the urban/suburban samples were collected from six different neighborhoods in Seattle, WA ($n = 120$). For the rural samples, the WDE reported a mean TEQ₂₀₀₆ concentration of 1.7 ng/kg (min–max: 0.1–9.4 ng/kg TEQ₂₀₀₆). It is important to note that the samples were collected in areas believed to be untouched by potential human or natural dioxin sources, including forest fires (WDE, 2011a). For samples collected in the six Seattle neighborhoods, the WDE reported a mean of 19.2 ng/kg TEQ₂₀₀₆ (min–max: 1.7–120 ng/kg TEQ₂₀₀₆). This is one of the few dioxin soil sampling efforts for which preliminary observations on temporal changes might be made. Although these data may suggest a very slight decrease (rural datasets) or increase (urban/suburban datasets) in soil dioxin levels since the 1999 WDE soil survey, the sampling efforts from these two different time periods did not overlap in specific location, and the number of sampling areas was much more substantial in the more recent survey. Therefore, no direct conclusions about the change in soil TEQ levels should be drawn from these datasets at this time.

3.15. Hawaii Soil Dioxin Contamination Report (HDOH, 2011)

The Hazard Evaluation and Emergency Response Office within the Hawaii Department of Health (HDOH) issued a report containing data on background concentrations of DLCs in Hawaiian soils (HDOH, 2011). The overall purpose of the study was to evaluate pesticide-

related contamination of soils in areas of former sugarcane operations. In order to determine if the contamination was in fact related to the sugarcane operations, HDOH also collected background samples from nine sites that were not associated with sugarcane operations. Of these nine sites, three of these sites were determined to best represent background soil. These three sites did not have a rural or urban/suburban designation by the original study authors, but they were classified as rural in this review based on assessment the site descriptions provided by the original study authors. The original study report only provided data on total TEQ concentrations. Data concerning concentrations of individual congeners was not provided for this study. For the three sites that were determined to be representative of background soils by the HDOH, the TEQ concentration ranged from 0.85 to 27 ng/kg (HDOH, 2011). The HDOH report did not investigate the potential impact of volcanic emissions on Hawaiian soil dioxin levels. Low levels of highly chlorinated PCDDs have been measured in volcanic ash (Takizawa et al., 1994; Feshin et al., 2006), therefore the constituent volcanic activity in Hawaii may contribute to background soil dioxin levels to some degree. It is important to note that the HDOH study employed a "multi-increment" (MI) sampling approach rather than the more common "discrete" sampling approach. The original study authors noted that the MI approach generates higher quality composite data than the discrete method, and based on previous experience, may result in higher soil dioxin measurements. Notably, HDOH indicated that most of the mainland background soil surveys likely underestimate actual dioxin concentrations, and that the USEPA is currently using the MI approach to re-investigate some dioxin-contaminated sites (HDOH, 2011).

3.16. USEPA Dioxin Reassessment (USEPA, 2003)

There are three additional unpublished soil surveys described in the 2003 USEPA Dioxin Reassessment for which the primary reports were unavailable. Two of these studies describe background dioxin concentrations in rural soils from Connecticut (MRI, 1992) and Maine (Tewhey Associates, 1997), with mean concentrations of 5.7 and 2.9 ng/kg TEQ₁₉₉₈, respectively. Additionally, the USEPA (2003) reported that NIH conducted a survey of the soils at their headquarters campus in Bethesda, MD and found a mean concentration of 2.2 ng/kg TEQ₁₉₉₈ across all samples. The USEPA (2003) designated the NIH samples as representative of urban background.

3.17. Additional background source information

In addition to the reports and publications described above, further information on concentrations of DLCs in background soils was collected through direct contact with several state agencies, either via telephone or email. Risk assessors at the Florida Department of Environmental Protection (FDEP) indicated that they have overseen the collection of soils for purposes of establishing site-specific background dioxin concentrations as a part of the investigation of Superfund sites within the state (FDEP, 2011). According to FDEP, background dioxin soil concentrations in Florida were typically found to range from 2 to 4 ng/kg. For the Escambia Superfund Site Assessment, where dioxin was a primary chemical of concern, background soil samples collected from non-contaminated soil were found to have TEQ₁₉₉₈ concentrations range from 0.3 to 9.5 ng/kg. The Massachusetts Department of Environmental Protection (MassDEP) indicated that their background screening level for residential soils (20 ng/kg TEQ) was not based on state-specific soil sampling (Swanson and Lamie, 2007), but rather represented the 90th percentile of the residential background soil concentrations reported as a part of the Denver, CO soil survey conducted by USEPA Region 8 (described above, USEPA, 2002). A small background soil dioxin study was conducted in the town of Canton, MA, but the raw data for this study were not readily available. Based on information in a secondary source (Swanson and Lamie, 2007), eight samples were taken at various

residential locations throughout Canton and analyzed for TCDD only. The average background TCDD concentration in these eight samples was reported to be 62 ng/kg, based on a range of 16–189 ng/kg. Additionally, in 1999 the Michigan Department of Environmental Quality (MDEQ) conducted a statewide survey of soil PCDD/F levels, reporting a range of 0.4–35.0 ng/kg TEQ₁₉₉₈ from 68 soil samples collected at various sites in both the upper and lower peninsulas (MDEQ, 1999). Although the sampling sites were reported to represent a mix of rural and urban areas, information released publicly was limited to a map with concentrations indicated but the resolution was not fine enough to be able to determine the specific land use in the area where the sample was collected, nor did the study authors provide summary statistics for rural vs. urban or other land uses.

4. Discussion

The results of this evaluation characterizing background concentrations of DLCs in urban/suburban and rural soils in the U.S. indicate that there is substantial variability in the concentrations reported in the literature. As demonstrated during the course of this current evaluation, there are also substantial differences in quality and design of the various studies—they did not all analyze for the same congeners (many studies did not account for DL-PCBs, and some studies only considered a few of the 17 PCDD/F congeners), they used different methods to impute values for non-detect results, they had different detection and reporting limits, they applied different TEFs (i.e., I-TEQ, WHO₁₉₉₈ TEF values, and WHO₂₀₀₆ TEF values) to calculate the total TEQ, land-use designations were not always clear and assumptions had to be made, etc. Because of these differences, not all of the data are directly comparable. That noted, and with some additional data evaluation, the data are still useful for gaining an understanding of the potential differences in concentrations across different land-uses and regions. While the USEPA has conducted a relatively comprehensive assessment of background concentrations in rural soils, no such assessment has been conducted for urban soils or soils in any other land-use categories. The data described herein indicate that the available data for rural soils is generally more robust than for urban/suburban soils. Additionally, and not surprisingly, background concentrations in urban/suburban soils tended to be higher and more variable than in rural soils, with maximum concentrations ranging from approximately 7–186 ng/kg on a TEQ basis in urban/suburban soils (means ranged from 2.2 to 51.2 ng/kg) and from about 2 to 23 ng/kg in rural soils (means ranged from 1.1 to 7.1 ng/kg). Since many of the datasets did not include a full DLC congener analysis, these ranges most likely represent an underestimation of the actual background TEQ concentrations. These findings demonstrate the importance of understanding background concentrations in soils across various land use types within the U.S., especially in light of the now substantially lower PRGs that have been developed by the USEPA and the possibility that these PRGs may drop even more once the Agency releases its updated cancer slope factor for TCDD.

The State of Hawaii's Department of Health (HDOH) clearly understood the importance of having a good characterization of background concentrations in soils when they recently established action levels for DLCs in soils (i.e., 240 ng/kg TEQ for residential soils and 1500 ng/kg TEQ for commercial/industrial sites) (HDOH, 2010). In establishing these levels, the State of Hawaii acknowledged that remediation of large tracts of agricultural lands where trace levels of dioxins associated with the past use of pentachlorophenol and other agricultural practices were identified as impractical and unnecessary from a health perspective. Also notable, Hawaii considers soils with 20 ng/kg TEQ or less to be natural background, and not to have been impacted by local, agricultural, or industrial releases of dioxins. Hawaiian soils with concentrations between 20 and 240 ng/kg TEQ are considered by HDOH to be minimally impacted by anthropogenic sources, citing that levels measured in soils from former agricultural fields ranged from b20 ng/kg to 100 ng/kg, and even up to 200 ng/kg in some areas. The HDOH also

considered typical dietary intake of dioxins with respect to theoretical risks posed by exposure to soils when establishing the values, and specifically discussed the relevance of USEPA's recently finalized TCDD RfD of 0.7 pg/kg-day (USEPA, 2010, 2012), noting that dietary intake of dioxins was estimated to exceed the hypothetical dioxin intake associated with long-term exposure to soils with TEQ concentrations at or below RfD-based action levels (HDOH, 2010). Based on this, HDOH concluded that it was impractical to use TCDD toxicity factors such as the current USEPA RfD value that equate to an exposure below dietary intake estimates, or background soil levels, to derive soil action levels.

The approach taken by the HDOH is of particular interest given that the USEPA had previously proposed interim PRGs (USEPA, 2009a). Currently, the USEPA recommends the derivation of site-specific cleanup levels using the current non-cancer-based toxicity benchmark that lead to cleanup values significantly lower (N90%) than the cleanup level that has been implemented for decades at CERCLA sites in the U.S. For the majority of Superfund sites in which DLCs have been identified as a chemical of concern, remediation goals have been set at 1000 ng/kg. Adoption of more stringent risk-based soil screening and cleanup criteria for DLCs may result in the reevaluation and reopening of Superfund sites where DLCs were not originally considered to present an unacceptable health risk, or where considerable resources have already been spent in site remediation (e.g., Times Beach, MO; Love Canal, Niagara Falls, NY; Pensacola, Escambia County, FL). These actions will likely result in unnecessary and expensive soil sample planning, collection and chemical analysis to characterize background. As acknowledged by the USEPA, because the draft PRGs would likely or possibly be below background concentrations of dioxins in U.S. soils, soil background levels would need to be characterized at most all sites.

The development of PRGs that are lower than or very close to background levels is additionally perplexing given that the majority of exposure (~90%) to DLCs in the general population is from diet (beef, pork, poultry, other meat, fish, milk, dairy, eggs), whereas less than 10% comes from exposure to DLCs in other sources (water, inhalation of air, ingestion of soil, soil dermal contact, and vegetable fat intake) (Lorber et al., 2009). In fact, several studies have demonstrated that long-term residence on soils with elevated levels of dioxins in soils has little, if any, impact on body burden (Ewers et al., 1996; ATSDR, 2005; Garabrant et al., 2009a; Hedgeman et al., 2009; Diliberto et al., 2008; Stehr-Green et al., 1988; Kimbrough, 1995; Kreiss, 1985; Yaffe and Reeder, 1989; Tohyama et al., 2011). Generally, the findings of these studies are consistent in that serum PCB or PCDD/F levels were similar to typical background levels despite exposures to dioxins in soils at concentrations above background (and well above background in some scenarios). Most recently, a study evaluating exposures to dioxins in soils from a residential area in Tokyo (Tohyama et al., 2011) reported a lack of association between body burdens and frequent exposures to contaminated soils at levels up to ~7000 ng/kg TEQ. The study included children, the receptor age group most likely to be exposed to soils, and demonstrated a lack of concordance of congener profiles between the soil and serum. Further, serum levels for residents from the high TEQ soils were similar to the general Japanese population. The only clear association reported was increased serum levels in infants exposed to dioxins via breastfeeding.

One of the largest-scale, most robust dioxin assessments conducted to date was the UMDES survey for Midland, MI, an evaluation of the potential relationship between the presence of DLCs in environmental media and residential serum (Garabrant et al., 2009b). No correlation between soil and serum DLC levels was observed (Garabrant et al., 2009a). Soil DLCs in the area believed to be impacted by a former industrial incinerator had the highest median concentration (58.2 ng/kg) of the areas examined; however, residents from the area population had the lowest median serum dioxin TEQ levels of any of the populations examined, including the control group (Hedgeman et al., 2009). Closer examination of participants who lived on land with the highest maximum soil concentrations (defined as N703 ng/kg) relative to those with lower

maximum soil concentrations (b703 ng/kg) indicated that most of the maximal soil concentration residents had serum TEQ concentrations well within the distribution of the other residents of the same age. The UMDES research team also developed linear regression models to identify significant predictors of \log_{10} serum dioxin concentrations. Soil and house dust dioxin concentration did not explain any of the variation in serum dioxin TEQ concentration; rather variations were explained best by demographic factors, in particular age (Garabrant et al., 2009a). These findings collectively demonstrated that contact with soils and house dust containing the elevated DLC levels present in the Midland area did not significantly impact background human body burdens. It is notable that, as part of its review of the UMDES study results, the USEPA conducted its own pharmacokinetic modeling to evaluate the impact of exposure to soils containing DLCs on body burden (USEPA, 2009b). The Agency reported that exposures to soils with elevated TEQ might not significantly affect adult body burdens. Specifically, data provided in the USEPA report on the UMDES study demonstrated limited impact for exposures of 1,000 ng/kg or less.

The application of the soil PRGs recently developed by the USEPA – levels that are have been shown often to be lower than background levels measured throughout the U.S. – would likely result in excessive and unnecessary remediation given the wealth of data indicating that exposures to DLCs in soils do not impact human body burdens. As noted by Kimbrough et al. (2010), such actions would likely not provide any health protective benefit. Further, the application of the new USEPA soil PRGs could lead to the conclusion that background soils in the U.S. may contain DLCs at unacceptable levels; such a conclusion could lead to unwarranted alarm and costly, unnecessary soil cleanup efforts. These concepts were directly addressed by the lead UMDES investigators in their comments regarding USEPA's draft interim PRG values (UMDES, 2010). Using data collected from their study, they commented that the proposed interim USEPA soil PRGs were not supported by site-specific data, and that establishing a PRG at 72 ng/kg or lower would result in the investigation of a substantial number of residential properties in areas with no known sources, including properties in the control region. In addition, USEPA has implemented a contradictory policy on dioxins where indirect soil exposure exceeding 0.7 pg/kg/day is held as unsafe, while the same degree of direct dietary exposure has been characterized as safe, including even much higher levels of breast milk TEQ exposure to infants (Lorber and Phillips, 2002) who are more susceptible and for whom the current RfD directly applies.

In conclusion, the findings of the assessment described herein demonstrate that there are considerable variations in background levels of DLCs in soils across the U.S. When considered in the context of several critical exposure and risk-related facts (declining DLC body burdens, diet as the most relevant DLC exposure route, and the minimal impact of soil exposures on human DLC body burdens), it seems impractical to establish regulatory remediation values for DLCs in soils that are likely to be below background concentrations. Going forward, it will be even more important to fully understand how proposed soil dioxin cleanup values, especially those based on the latest TCDD toxicity criteria, compare to background concentrations in urban, suburban, agricultural, and rural soils — and the impact of such comparisons, particularly for evaluation of Superfund sites. The background data summarized in this paper indicates the need for more robust characterizations of urban, suburban, and agricultural soils — particularly in light of the pending updates to regulatory values. The variability across datasets is the result of disparate sampling studies and objectives conducted at various timepoints over the previous three decades, and highlights the need for a robust soil study that encompasses rural, suburban, and urban areas.

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